Impact of top-down isoprene emissions on



surface ozone over the Amazon Basin



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1. Introduction

- Isoprene is the dominant biogenic volatile organic compound emitted by terrestrial vegetation, and an important precursor of tropospheric ozone.
- The goal of my research is to use satellite measurements of formaldehyde (HCHO) vertical columns, retrieved by the SCIAMACHY and OMI instruments, to quantify isoprene emissions from the Amazon rainforest (the largest source of isoprene into the atmosphere).
- To achieve this objective, I've been developing a nest-grid version of the GEOS-Chem chemistry transport model, centred over the Amazon, to interpret the satellite data.
- I've run multiple model simulations to determine an ensemble of top-down isoprene emissions to better characterise the uncertainties. This poster shows some of the results and the subsequent impact of the top-down emissions on surface ozone.

2. Overview of instruments and GEOS-Chem configurations

Table 1: Summary of the SCIAMACHY and OMI instruments and their HCHO retrievals.

| | SCIAMACHY | OMI |
|---------------------------------|---|--|
| Instrument | | |
| Platform | ENVISAT | EOS-Aura |
| Orbit | Sun-synchronous, descending node | Sun-synchronous, ascending node |
| Local Equator crossing time | 10:00 | 13:30 |
| Swath | 960 km | 2600 km |
| Main observing mode | Alternating limb/nadir sequence | Continuous nadir |
| Pixel size (at nadir) | $30\times60~\mathrm{km}^2$ | $13 \times 24 \text{ km}^2$ |
| Global coverage | 6 days | 1 day |
| Spectral range | 220–2380 nm | 270–500 nm |
| HCHO retrieval | | |
| Fitting window | 327.5–356.5 nm | 328.5–346 nm |
| Spectral resolution (at 340 nm) | 0.26 nm | 0.42 nm |
| Fitting method | DOAS | Direct radiance fitting |
| Fitted species | HCHO, O ₃ , O ₄ , NO ₂ , BrO, Ring, OCIO | HCHO, O ₃ , NO ₂ , BrO, Ring |

| Table 2: Summary of the different GEOS-Chem scenarios used to infer top-down isoprene emissions from SCIAMACHY and OMI. | | |
|---|--|--|
| Scenario | Description | |
| PCEEA HYBRID | Default scenario for each instrument. Isoprene emissions calculated using the PCEEA algorithm of Guenther et al. [2006] Isoprene emissions calculated using a 5-layer canopy model and a combination of Guenther et al. [1995,2006] algorithms | |
| MULLER | As HYBRID, but with isoprene emissions scaled by 0.635 to match emissions from the study of Muller et al. [2008] | |
| LPJ-G5 | Emissions based on the LPJ-GUESS model forced with GEOS-Chem's GEOS-5 meteorology | |
| LPJ-CRU | Emissions based on the LPJ-GUESS model forced with its default CRU meteorology | |
| BL | As the default scenario but using a non-local boundary layer mixing scheme | |
| SLOWDEP | As the default scenario but without the fast deposition of oxygenated VOCs | |
| HPALD | As the default scenario but assuming fast photolysis of hydroperoxy aldehydes | |
| LIM0 | As the default scenario but with the explicit treatment of hydroperoxy aldehydes | |
| KPP | As the default scenario but using the KPP chemical solver in GEOS-Chem | |
| CHEMT | As the default scenario but using a 10 min emissions and chemistry time step (instead of 60 min) | |
| ALB | As the default scenario but using the surface reflectances in the AMF computation | |
| CF+ | As the default scenario, but assuming a ± 0.1 cloud fraction error in the AMF computation | |
| CF- | As the default scenario, but assuming a -0.1 cloud fraction error in the AMF computation | |
| CTP+ | As the default scenario, but assuming a $+60$ hPa error in cloud top pressure in the AMF computation | |

3. Top-down isoprene emissions

As the default scenario, but assuming a -60 hPa error in cloud top pressure in the AMF computation

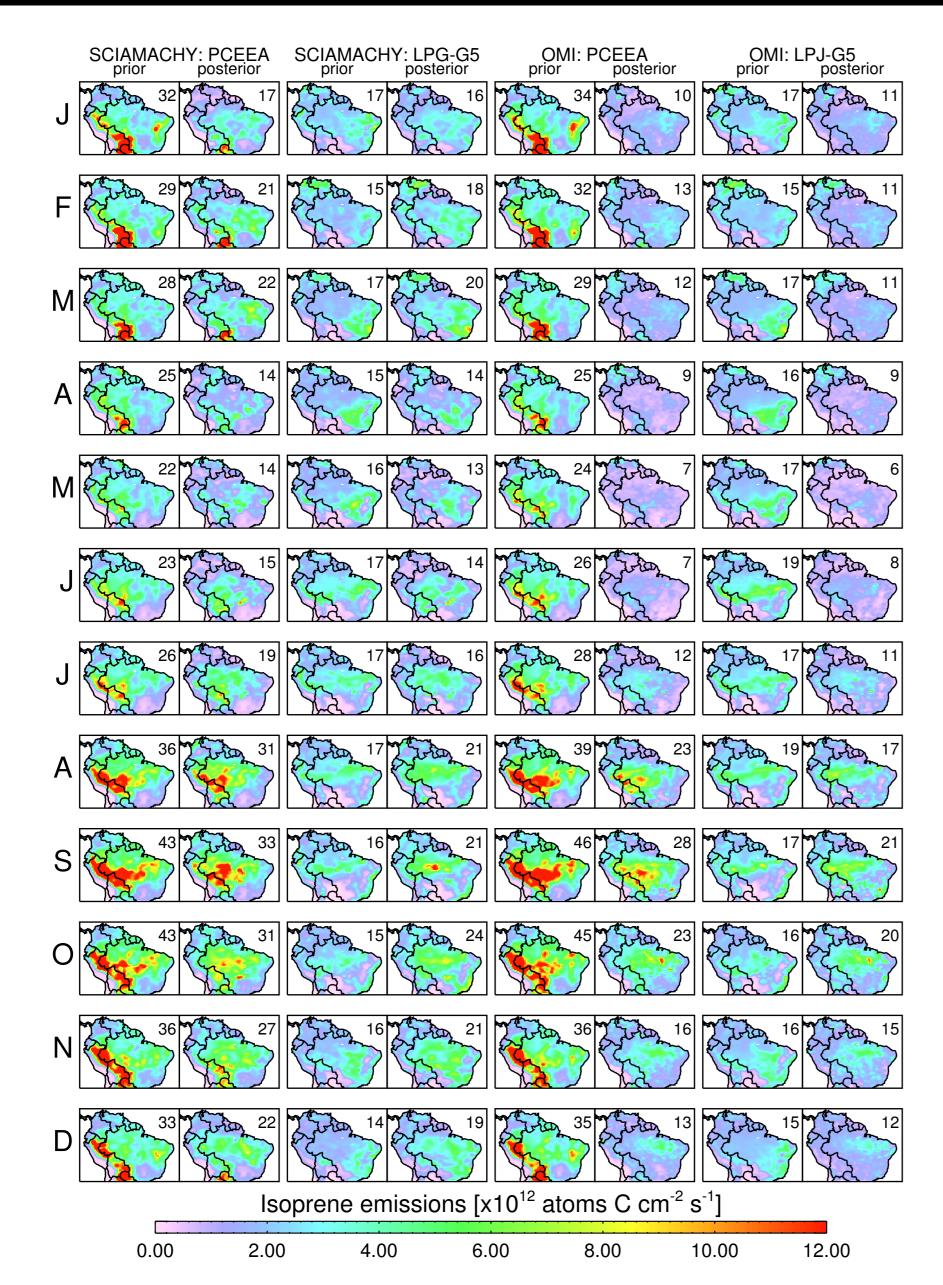


Figure 1: Contiguous maps of prior and posterior isoprene emissions corresponding to the overpasses of SCIAMACHY (9-11 LT) and OMI (12-15 LT) using the PCEEA and LPJ-G5 scenarios . The emissions have been smoothed with a 3×3 box-filter (for illustration only); the monthly totals in Tg C are shown inset. Within our modelling framework, we find use of the HCHO column data more tightly constrains the ensemble isoprene emission range from 193-393 Tg C to 217-292 Tg C for SCIAMACHY, and 201-442 Tg C to 149-197 Tg C for OMI. Median uncertainties of the top-down emissions are about 70–105% for SCIAMACHY, and 50–90% for OMI. We find the inferred emissions are most sensitive to the choice of chemical solver, uncertainties in cloud fraction and cloud top pressure, the initial bottom-up isoprene emission inventory used, and the retrieval of the HCHO vertical column itself.

4. Impact of top-down emissions on simulated HCHO columns

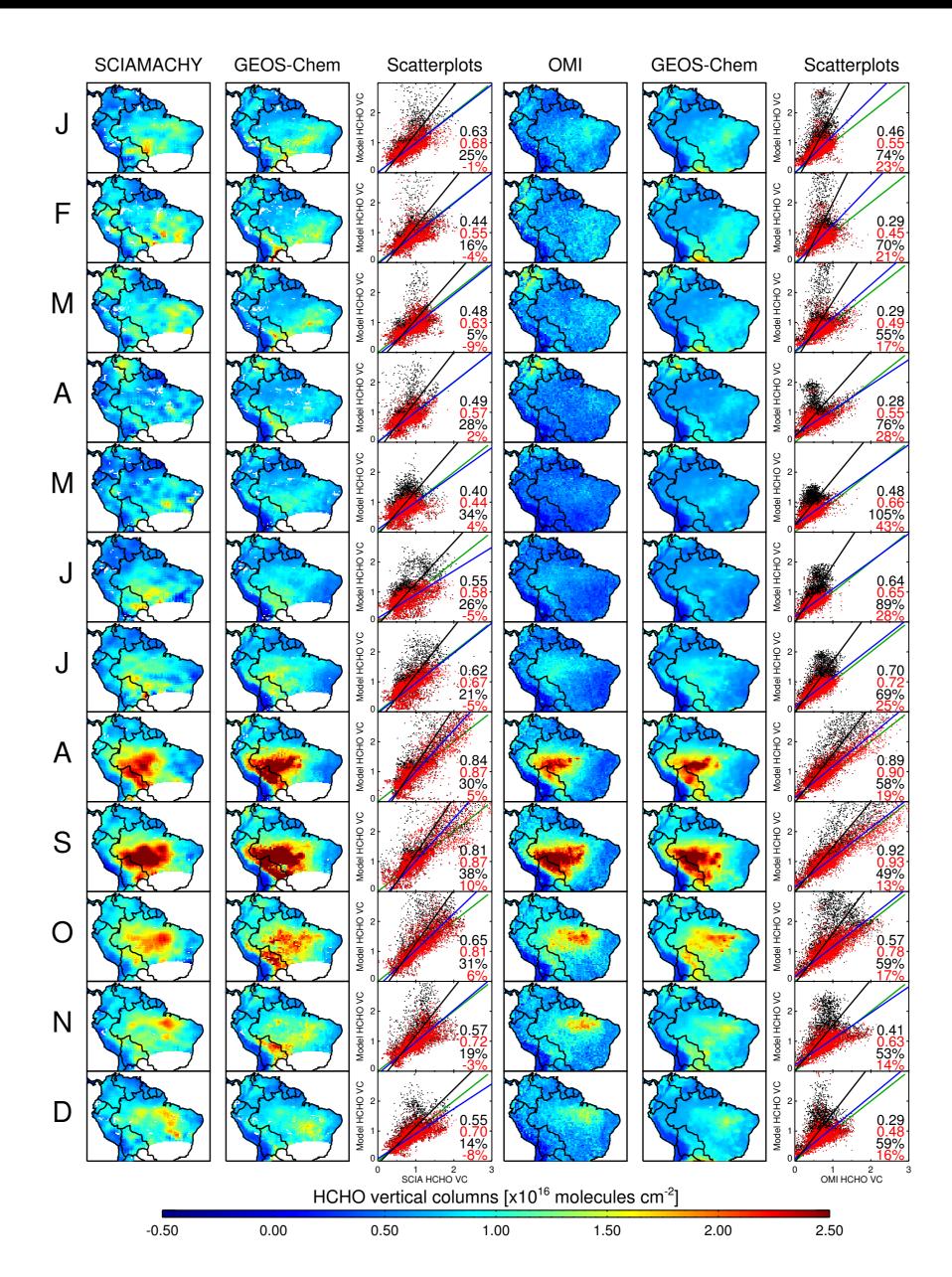


Figure 2: The first two columns show SCIAMACHY and GEOS-Chem HCHO vertical columns, when the model has been constrained by both SCIAMACHY (at 9-11 LT) and OMI (at 12-15 LT) top-down emissions. The fourth and fifth columns show OMI and GEOS-Chem HCHO vertical columns, when the model has been constrained at all time-steps by OMI top-down emissions alone. The corresponding scatterplots show SCIAMACHY and OMI versus GEOS-Chem HCHO vertical columns, that have been simulated using (1) the original prior PCEEA emissions (black dots; black line is reduced major axis fit) and (2) the inferred top-down emissions (red dots; blue line is regression fit). The x=y line is in green. The normalised mean bias and Pearson correlation coefficients are given inset.

5. Impact of top-down emissions on simulated surface ozone

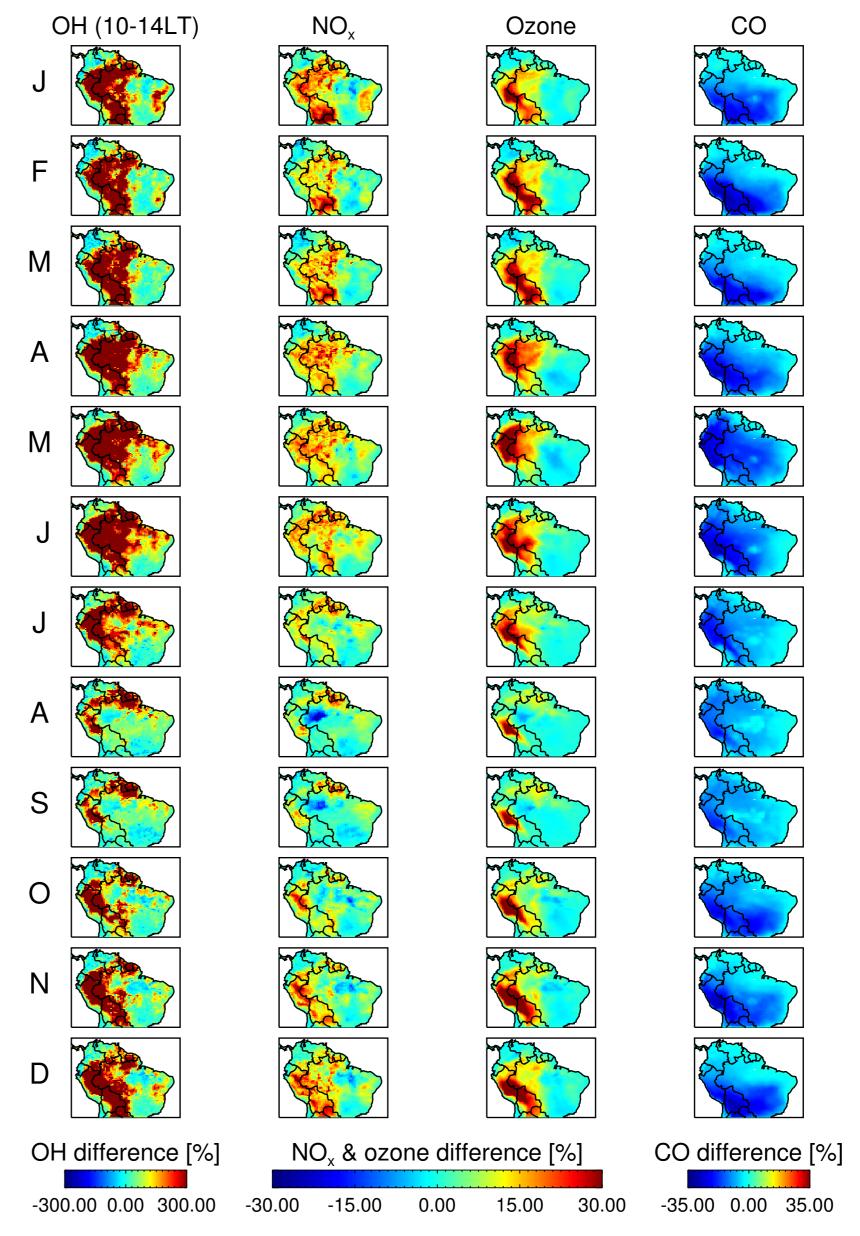


Figure 3: The effect on GEOS-Chem monthly mean surface concentrations of OH, NO, ozone and CO, simulated using top-down emissions from the OMI/PCEEA scenario. Difference = $100\% \times (posterior - prior) / prior$. Surface ozone concentrations change from 8-72 ppbv (prior emissions) to 11-70 ppbv (posterior emissions), i.e. minimum surface ozone values are increased but maximum values are decreased.

6. Summary

- Top-down isoprene emissions inferred from SCIAMACHY and OMI improve GEOS-Chem's simulation of HCHO over the Amazon
- The impact of the top-down emissions in surface ozone concentrations is moderate; the impact on OH is substantial.
- Future work will compare GEOS-Chem ozone to SHADOZ profiles to verify if the model ozone simulation is truly improved.